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Adam J. Trevitt
University of Melbourne, adamt@uow.edu.au

Philip J. Wearne
University of Melbourne

Evan J. Bieske
University of Melbourne

Michael D. Schuder
Carroll College - Waukesha

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Abstract

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Observation of nondegenerate cavity modes for a distorted polystyrene microsphere

Adam J. Trevitt, Philip J. Wearne, and Evan J. Bieske

School of Chemistry, The University of Melbourne, 3010, Parkville, Victoria, Australia

Michael D. Schuder

Department of Chemistry, Carroll College, Waukesha, Wisconsin 53186

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Nondegenerate azimuthal morphology-dependent resonances are observed for a distorted, fluorescently labeled polystyrene microsphere levitated in a quadrupole ion trap. Modeling the individual resonances by using perturbation theory allows a determination of quadrupole and octupole distortion parameters. The particle's shape changes slowly over the course of the measurement and eventually becomes spherical. The morphological changes are facilitated by laser heating of the particle above the polystyrene glass transition temperature. We demonstrate a method of transforming a trapped particle to a sphere and rendering its azimuthal modes degenerate. © 2006 Optical Society of America

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It is well known that a microsphere can act as a three-dimensional optical cavity exhibiting morphology dependent resonances (MDRs) or whispering gallery modes.[1–4] The MDRs of a dielectric sphere, which are distinguished by polarization (TE or TM), mode order ν, and angular momentum number l, have $2l+1$ azimuthal degeneracy. The degeneracy is removed when the sphere is distorted to an ellipsoid so that for a given polarization, ν, and l, the system exhibits $(l+1)$ distinct azimuthal modes (labeled $m=l, l\text{-}1, \ldots, 0$). Optical effects resulting from shape distortion have been reported for droplets generated by a vibrating orifice where periodic distortions from sphericity occur on millisecond time scales.[5,6] In this Letter, we study the slow morphological changes of a single fluorescent polystyrene microparticle confined in a quadrupole ion trap (QIT). The shape parameters for the particle are extracted by modeling the splitting and shifting of nondegenerate azimuthal MDRs by using the perturbation theory developed by Lai *et al.*[7]

We report here a case in which nondegenerate azimuthal MDRs are spectrally resolved and assigned, thus providing an opportunity to test and apply current theoretical descriptions of distorted microsphere MDRs.[7] Furthermore, this work demonstrates that laser-induced melting can transform a trapped polystyrene particle into a sphere and render its azimuthal modes degenerate.

The QIT used in this study (shown in Fig. 1) is similar to that reported by Schlemmer *et al.*[8] and was operated in single-phase mode at 250 Hz frequency and 740 V amplitude. Fluorophore labeled polystyrene microspheres (Sigma-Alrich, nominal diameter 2.16 μm) were introduced into the QIT by using matrix-assisted laser desorption–ionization (MALDI), with 3-hydoxypicolinic acid (3-HPA) as the absorbing matrix. To prepare a MALDI sample, three to four drops of an aqueous suspension of the polystyrene particles were mixed with three to four drops of a saturated solution of 3-HPA in an acetonitrile–water mixture on a stainless steel sample tip. The sample was then dried under nitrogen and placed above the QIT situated in a vacuum chamber. A single 7 ns laser pulse, third harmonic ($\lambda=355$ nm) from a Q switched Nd:YAG laser, irradiated the sample, ejecting charged particles into the QIT. Usually the particles carried 1000–1400 elementary positive charges. By systematically varying the trap driving voltage and frequency, the particles were ejected until only one remained. By using He buffer gas ($\sim$100 mTorr) we damped the remaining particle's motion so that its oscillation amplitude was <0.1 mm. Approximately 2 mW from a continuous wave a 532 nm $Nd:YVO_4$ laser irradiated the trapped particle. Scattered or fluorescent light was collected perpendicular to the incident laser beam by using a 25 mm diameter $F/1$ lens and directed through an edge filter to remove 532 nm light and then into a spectrometer (with a resolution $\sim$0.03 nm) equipped with a CCD detector interfaced to a personal computer.

The fluorescence emission spectrum of most trapped particles was dominated by the MDR's characteristic of a dielectric sphere and could be modeled using Mie theory yielding the particle's radius (a_0) and refractive index (m_λ).[9] However, several particles exhibited more complex spectra indicating distortions from sphericity. Moreover, the particles ap-

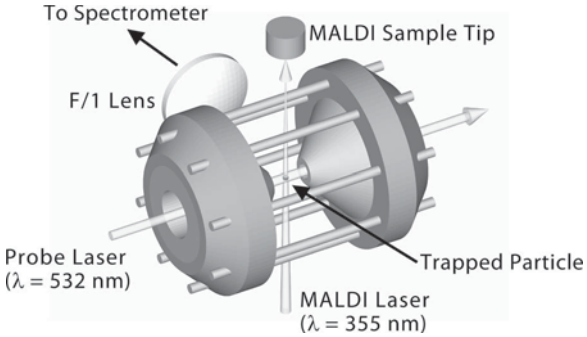

Fig. 1. View of the experimental configuration.

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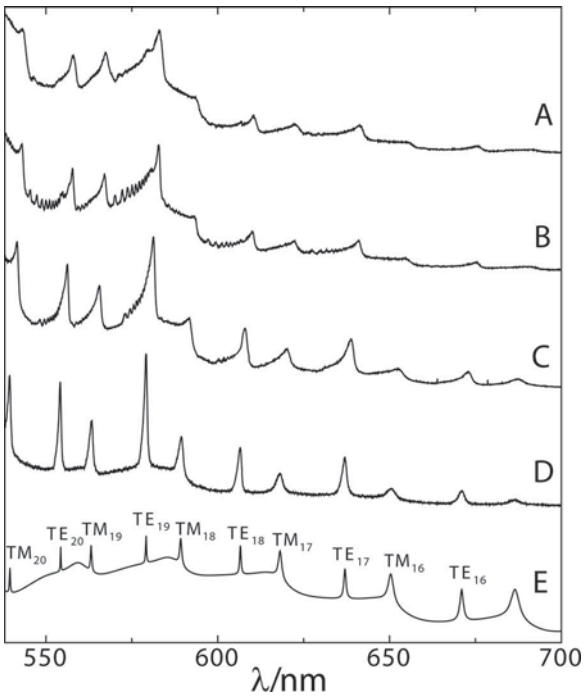

Fig. 2. A–D, Fluorescence spectra from a single polystyrene particle taken at ~5 min intervals. Spectral changes are due to the particle's relaxation from spheroidal to spherical shape. E is the predicted scattering spectrum for a sphere with $a_0 = 1363$ nm and $m_\lambda = 1.549 + 10{,}230\ \text{nm}^2/\lambda^2$.

peared to gradually relax to a spherical form. For example, Fig. 2 shows the consecutive spectra for a particle recorded at ~5 min intervals where the emission profile changes significantly over 20 min. Initially the spectrum displays several broad peaks tailing off to the blue (spectrum A). After some time small resolvable peaks become apparent to the blue of the TE_{20} and TE_{19} peaks (spectra B and C). These features are attributable to nondegenerate azimuthal modes associated with a nonspherical particle. After 20 min, the emission spectrum has become that of a spherical particle and the MDRs are modeled and labeled accordingly. The theoretical Mie scattering spectrum [Fig. 2(E)] was generated by assuming a particle $a_0 = 1363$ nm and $m_\lambda = 1.549 + 10230\ \text{nm}^2/\lambda^2$.

Nondegenerate azimuthal MDRs clearly resolved in traces B and C (Fig. 3) have wavelengths consistent with the particle's being a prolate spheroid. For a given l, the various m modes have different effective path lengths and hence different resonant wavelengths. The $m = l$ mode confined around the spheroid equator has the shortest wavelength, while the $m = 0$ mode confined to an ellipse passing through the poles has the longest wavelength.

To derive information on the particle's shape we follow Lai *et al.*,[7] who developed expressions for the MDR frequencies of a distorted dielectric sphere. They consider a sphere affected by an axially symmetric shape perturbation such that the radius is

$$r(\theta,\phi) = a + \Delta_L \sqrt{4\pi} Y_{L,0}(\theta,\phi), \tag{1}$$

where Δ_L is a weighting term for the $Y_{L,0}(\theta,\phi)$ spherical harmonic. The shift of the m modes ($\delta\omega$) with the same l relative to that of a sphere with radius a is given as

$$\frac{\delta\omega}{\omega} = -\frac{\Delta_L}{a} F(L,l,m), \tag{2}$$

where $F(L,l,m)$ is defined as

$$F(L,l,m) = A(L,l) f(m), \tag{3}$$

with

$$A(L,l) = \frac{2l+1}{\sqrt{2L+1}} [C(llL;000)]^2 \left[1 - \frac{L(L+1)}{2l(l+1)}\right], \tag{4}$$

$$f(m) = (-1)^m \frac{C(llL;m,-m,0)}{C(llL;000)}. \tag{5}$$

The $C(j_2 j_3; m_1 m_2 m_3)$ are Clebsch–Gordan coefficients.

For odd L distortions, $F(L,l,m)$ is zero and the m modes remain degenerate.[7] The lowest-order perturbation to remove the degeneracy is a quadrupole distortion $Y_{2,0}(\theta,\phi)$, which for $l \gg 1$ results in m-mode shifts described by

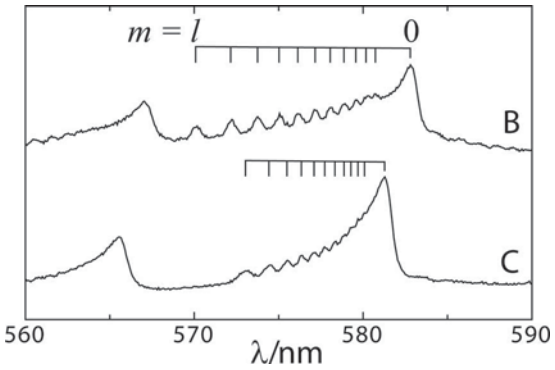

Fig. 3. Expanded sections of spectra B and C from Fig. 1. Nondegenerate azimuthal MDRs are labeled. The $m = l$ mode has the shortest wavelength and the $m = 0$ mode the longest, consistent with a prolate spheroidal particle.

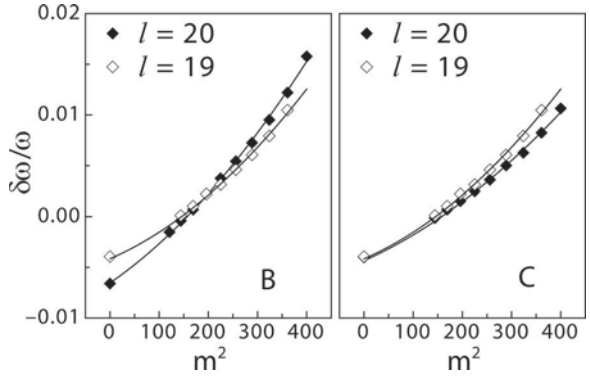

Fig. 4. Measured fractional frequency shifts for the TE_{20} and TE_{19} m modes in spectra B and C (points). Lines represent frequency shifts calculated by using Eq. (8) and the fitted Δ_2 and Δ_4 values in Table 1.

Table 1. Fitted Values of Quadrupole (Δ_2) and Octupole (Δ_4) Distortion Parameters[a]

Spectrum	l	$a\pm3$	$\Delta_2\pm0.2$	$\Delta_4\pm0.3$	$r_p\pm3$	$r_e\pm3$
B	20	1362	18.0	−2.6	1395	1339
	19	1362	17.8	−2.7	1394	1339
C	20	1362	12.0	−1.8	1384	1347
	19	1362	11.8	−2.1	1382	1347

[a]The fitted values are expressed in nanometers for TE_{19} and TE_{20} m modes in Spectra B and C. The spheroid's polar and equatorial radii (r_p and r_e in nanometers) are also listed.

$$\frac{\delta\omega}{\omega} = -\frac{\Delta_2}{a}F(2,l,m) = -\frac{\Delta_2}{a}\frac{\sqrt{5}}{4}\left[1 - \frac{3m^2}{l(l+1)}\right]. \quad (6)$$

As seen in Fig. 4, the relative m-mode displacements ($\delta\omega/\omega$) in spectra B and C display an almost quadratic dependence on m as predicted by Eq. (6). However, a small additional quartic term is necessary to fit the data. Thus an additional axial-symmetric $Y_{4,0}(\theta,\phi)$ perturbation was included to extend Eq. (1) to

$$r(\theta,\phi) = a + \Delta_2\sqrt{4\pi}Y_{2,0}(\theta,\phi) + \Delta_4\sqrt{4\pi}Y_{4,0}(\theta,\phi), \quad (7)$$

and Eq. (2) to

$$\frac{\delta\omega}{\omega} = -\frac{\Delta_2}{a}F(2,l,m) - \frac{\Delta_4}{a}F(4,l,m). \quad (8)$$

For $L=4$,

$$A(4,l) = \frac{27(l^2+l-10)(l+2)(l-1)}{4(2l-1)(2l+5)(4l^2-9)}, \quad (9)$$

$$f(m) = 1 - \frac{5(6l^2+6l-5)m^2}{3l(l^3+2l^2-l-2)} + \frac{35m^4}{3l(l^3+2l^2-l-2)}. \quad (10)$$

The m-mode frequencies in the TE_{20} and TE_{19} regions of spectra B and C were fitted to Eq. (8) to give Δ_2 and Δ_4 values (Table 1). The value of a was constrained by assuming that the particle's volume remained constant. The fitted and experimental m-mode displacements are in very good agreement (Fig. 4).

The foregoing analysis shows that the evolving emission spectra in Fig. 2 are consistent with the slow change of a particle's shape from prolate spheroid to sphere. The shape change, presumably driven by surface tension, suggests that the particle's temperature exceeded the polystyrene glass transition temperature (T_g), which for bulk material is ~373 K.[10] A particle temperature in the 400–450 K range can be arrived at through estimations of the heat input due to laser light absorption and heat loss due to black body radiation and buffer gas cooling.

The final radius of the sphere in Fig. 2(D) (1363 nm) is somewhat larger than the manufacturer's quoted radius for the polystytrene spheres (1080 nm). Tellingly, a sphere with a radius of 1361 nm has the same volume as two spheres with a radius of 1080 nm. Therefore it seems likely that the emission spectra in Figs. 2(A)–2(D) are the signatures of the final stages of two smaller spheres fusing into one.

In summary, we have shown that it is possible to laser melt an isolated microparticle suspended in a QIT, transforming it into a sphere, and rendering its azimuthal modes degenerate. The quantitative details of the spheroidal particle's shape have been extracted by assigning and modeling the spectrally resolved nondegenerate azimuthal modes.

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